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35 USC § 103(a) Rejections

The Examiner has rejected claims 1-56 under 35 USC § 103(a) as being unpatentable over **Buysch et al.** (US 5,856,554) and **Mizukami et al.** (US 5,380,907), or alternatively one of **Pressman et al.** (US 5,760,272; hereinafter referred to as **Pressman '272**) and **Pressman et al.** (US 6,114,564; hereinafter referred to as **Pressman '564**). The Applicants courteously traverse these rejections.

The Examiner states that (quote) :

- "The instant application claims a process for the production of diaryl carbonates, diphenyl carbonate, by the reaction of an aromatic hydroxy compound, a phenol, with carbon monoxide and oxygen in the presence of catalyst composition comprising a group 8, 9, or 10 metal, palladium, at least one salt, at least one metal co-catalyst, and optionally an activating solvent, and where the reaction is commenced at a temperature sufficient to keep the aromatic hydroxy compound molten. Further, the reaction is performed under various other conditions, such as when the temperature or pressure is increased, when the CO and O₂ is mixed, and when it is introduced, etc."

The Examiner further states that (quote) :

- "The prior art, **Buysch et al.**, teaches a process for the production of diaryl carbonates by the reaction of an aromatic hydroxy compound with CO and O₂ in the presence of a catalyst composition comprising platinum, a metal co-catalyst, a quaternary salt, and a base, and at temperatures and pressures that are within the scope of that instantly claimed. Further, the reference teaches that it is advantageous to conduct the process under conditions where the starting hydroxy compound is in the melt with diaryl carbonate, corresponding to the hydroxy compound, see col 2 line 42 thru col 3 line 16 and examples 1-6."

The Examiner goes on to state that **Buysch** does not teach that an activating solvent may be present or the preference for palladium in the catalyst composition, but

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that **Mizukami** or alternatively one of the **Pressman** references do teach that an activating solvent may be present and the preference for palladium in the catalyst composition.

The Examiner further states that (quote) :

- "The only difference of the combined references is that the **Buysch et al** reference teaches that DPC is present in the initial melt with the hydroxy compound. However, since this appears to be the only variation of the instant application from the bulk of the prior art, all other parameters being equal to that known in the art, it would have been obvious to combine the known art, and making slight variations in order to provide an optimized process. Further, given the results of the **Buysch et al** reference the results of the instant application show no unexpected results to indicate that the instant process is any better than the prior art, e.g. the prior art process produces yields of DPC that are 3 to 4 times that instantly taught."

The Applicants respectfully assert that, contrary to the Examiner's statement, **Buysch** does not produces yields of DPC that are 3 to 4 times that instantly taught. First of all, the DPC values presented in **Buysch** are wt. % values, not yield values. The process of **Buysch** requires that DPC be present in the reaction mixture before reaction of phenol with oxygen and carbon monoxide commences. Therefore, the wt. percentage of DPC present at any given time in **Buysch's** examples must be corrected for the amount of DPC present at the beginning of the reaction in order to determine how much new DPC has formed through the desired reaction of phenol with carbon monoxide and oxygen. More particularly, in **Buysch's** Example 1 (column 9, lines 8-12) it is stated that (quote): "The analyses showed that the reaction mixture contained 30.1% by weight diphenyl carbonate after one hour...." This value for % by weight DPC in Example 1 of **Buysch** has not been corrected by subtracting the amount of DPC initially added to the reaction mixture, namely 21.4% by weight. Similar language is present in all the other examples given in **Buysch**.

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To aid in the examination process the Applicants submit Table 1 below containing calculations of wt. % DPC for each example in **Buysch** taking into account the amount of DPC initially present in each reaction mixture. Values for "Wt. % DPC initially present" are already given in **Buysch** after the Example headings for Examples 1-4, and were calculated based on the combined weight of phenol and DPC initially present. Therefore, for Table 1 below the values for "Wt. % DPC initially present" for Examples 5-6 were similarly calculated based on the combined weight of phenol and DPC initially present.

TABLE 1. Actual Wt. % Values for DPC in **Buysch** Examples

Buysch Example	Wt. % DPC initially present	Wt. % DPC present after time period	Actual Wt. % new DPC	Wt. % new DPC <u>estimated</u> at 1.5 hr.
1	21.4	30.1 (1 hr.) 38.5 (2 hr.)	8.7 (1 hr.) 17.1 (2 hr.)	12.9
2	35.7	43.9 (1 hr.) 51.9 (2 hr.)	8.2 (1 hr.) 16.2 (2 hr.)	12.2
3	50	58.1 (1 hr.) 66.0 (2 hr.)	8.1 (1 hr.) 16.0 (2 hr.)	12.1
4	21.4 (same as Example 1)	28.3 (1 hr.) 35.1 (2 hr.)	6.9 (1 hr.) 13.7 (2 hr.)	10.3
5	20	28.2 (5 hr.)	8.2	---
6	50	58.7 (5 hr.)	8.7	---

Also included in Table 1 are values for wt. % DPC in **Buysch** estimated at 1.5 hours (calculated by dividing by 2 the sum of wt.% values at 1 hour and at 2 hours). These estimated values are included for easier comparison to corresponding wt.% DPC values in Tables 1 and 2 of the instant application, all of which were taken at 1.5 hours. In the instant application it can be seen that the range of wt. % DPC values in Tables 1 and 2 (Examples 1 and 6-11 in Table 1 and Examples 12-15 in Table 2) is 14.2-18.0, all of which values are higher than the estimated values in **Buysch** taken at the same time period. In fact, a number of wt. % DPC values at 1.5 hours reaction time in Tables 1 and 2 of the instant application are higher than those reported in **Buysch** at 2 hours and even 5 hours reaction time.

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The Applicants respectfully submit that in one of its embodiments the present invention is directed to an effective start-up procedure for a carbonylation reaction. Among other advantages this effective start-up procedure avoids rapid inactivation of carbonylation catalyst system and results in high yields of aromatic carbonate (kindly see the instant application paragraph 47, lines 1-4). If wt. % values for diphenyl carbonate are appropriate for comparison as the Examiner appears to suggest, then in a particular embodiment the advantages of the Applicants' invention are reflected in the improved wt. % values for diphenyl carbonate obtained early in the reaction course compared to those in **Buysch**.

Considerations of yield aside, the teachings of **Buysch** are directed to a process for making diaryl carbonate, which process requires the presence of at least 20 wt. % of said diaryl carbonate in the reaction mixture at the start of the reaction (kindly see **Buysch**, claim 1). In contrast the claims of the instant invention have no requirement that diaryl carbonate be present at the start of the reaction, and achieve excellent results without said presence. The Applicants respectfully submit that there is no motivation based on the teachings of **Buysch** for anyone skilled in the art to perform a process to make diaryl carbonate in the absence of diaryl carbonate present initially in the reaction mixture. Furthermore, there is no guidance in the teachings of **Buysch** for one skilled in the art to perform a process to make diaryl carbonate in the absence of diaryl carbonate present initially in the reaction mixture.

The process taught in **Buysch** also requires that a metal co-catalyst (described in **Buysch**, column 5, lines 63-67 through column 6, lines 1-14) be added after the other reaction components have been treated with carbon monoxide (kindly see **Buysch**, Examples 1-4) or else that no carbon monoxide treatment of the reaction mixture take place before treatment with a carbon monoxide-oxygen mixture (kindly see **Buysch**, Examples 5 and 6). In contrast, the claims of the instant invention require that reaction components, including metal co-catalyst, be treated with carbon monoxide before treatment with a carbon monoxide-oxygen mixture. The Applicants respectfully submit that there is no motivation based on the teachings of **Buysch** for anyone skilled in the art to perform a process to make diaryl carbonate which involves treatment of a reaction

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mixture comprising a metal co-catalyst with carbon monoxide before treatment with a carbon monoxide-oxygen mixture. Furthermore, there is no guidance in the teachings of **Buysch** for one skilled in the art to perform a process to make diaryl carbonate which involves treatment of a reaction mixture comprising a metal co-catalyst with carbon monoxide before treatment with a carbon monoxide-oxygen mixture.

Turning now to **Mizukami**, the Applicants respectfully submit that there is no teaching or suggestion in **Mizukami** of the use of a salt as a reaction component. In contrast, the claims of the instant invention require the use of at least one salt. The Applicants respectfully submit that there is no motivation based on the teachings of **Mizukami** for anyone skilled in the art to perform a process to make diaryl carbonate in the presence of a salt as a required reaction component. Furthermore, there is no guidance in the teachings of **Mizukami** for one skilled in the art to perform a process to make diaryl carbonate in the presence of a salt as a required reaction component.

Turning now to **Pressman '272** and **Pressman '564**, the Applicants respectfully submit that there is no teaching or suggestion in either **Pressman '272** or **Pressman '564** of carbon monoxide treatment of the reaction mixture before treatment with a carbon monoxide-oxygen mixture (kindly see **Pressman '272**, Examples 1 and 2, and **Pressman '564**, Examples 1-16). In contrast, the claims of the instant invention require carbon monoxide treatment of the reaction mixture before treatment with a carbon monoxide-oxygen mixture. The Applicants respectfully submit that there is no motivation based on the teachings of either **Pressman '272** or **Pressman '564** for anyone skilled in the art to perform a process to make diaryl carbonate which involves treatment of a reaction mixture with carbon monoxide before treatment with a carbon monoxide-oxygen mixture. Furthermore, there is no guidance in the teachings of either **Pressman '272** or **Pressman '564** for one skilled in the art to perform a process to make diaryl carbonate which involves treatment of a reaction mixture with carbon monoxide before treatment with a carbon monoxide-oxygen mixture.

The Examiner further states that (quote) :

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- "Therefore, one would have been motivated to combine the references to arrive at the instantly claimed process since the various catalyst compositions are well known in the art for the production of diaryl carbonates, and to provide an optimized process. Thus, it would have been obvious to one of ordinary skill in the art ... to combine the references and to modify the prior art given the state of the art at the time the invention was made in order to attempt to optimize the process absent the showing of unexpected results."

Considering the combination of **Buysch** and **Mizukami**, or alternatively one of **Pressman '272** and **Pressman '564**, the Applicants respectfully submit that combining **Buysch** with **Mizukami**, or alternatively one of **Pressman '272** and **Pressman '564**, does not give the Applicants' invention. **Buysch** requires the presence of diaryl carbonate product in the reaction mixture before commencement of reaction of a phenol with carbon monoxide and oxygen. One advantage of the present invention is that the presence of diaryl carbonate product in the reaction mixture is not required before commencement of reaction of a phenol with carbon monoxide and oxygen. Also, **Buysch** requires that a metal co-catalyst be added after the other reaction components have been treated with carbon monoxide or else that no carbon monoxide treatment of the reaction mixture take place before treatment with a carbon monoxide-oxygen mixture. Another advantage of the present invention is that reaction components, including metal co-catalyst, are treated with carbon monoxide before treatment with a carbon monoxide-oxygen mixture.

The Applicants respectfully submit that the Examiner's statement that (quote):

- "...it would have been obvious ... to attempt to optimize the process (of diaryl carbonate formation)...." (emphasis added)

is an admission that there is merely a "suggestion to try" the combining of process elements taken randomly from **Buysch** and **Mizukami**, or alternatively one of **Pressman '272** and **Pressman '564**, and with the requirement that critical process elements be omitted from **Buysch's** process. There are no examples or guidance in any of these

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references that might aid one skilled in the art in implementing a process of the instant invention.

The Applicants further note that there is no teaching or suggestion in **Buysch**, **Mizukami**, **Pressman '272** or **Pressman '564** or in any combination thereof wherein a reaction mixture is heated at atmospheric pressure to a temperature in a range of between about 72°C and about 90°C; then pressurized with carbon monoxide as required in steps (ii) and (iii) of claim 33 of the instant application. More particularly, in **Buysch** in Examples where there is a treatment with carbon monoxide before treatment with carbon monoxide-oxygen mixture, the reaction mixture is heated to 60°C (kindly see **Buysch**, Examples 1-4). In Examples in **Mizukami**, the reaction mixture is treated at room temperature (kindly see **Mizukami**, Examples 1-18). In Examples in **Pressman '272** and **Pressman '564** there is no treatment with carbon monoxide before treatment with carbon monoxide-oxygen mixture.

The Applicants further note that there is no teaching or suggestion in **Buysch**, **Mizukami**, **Pressman '272** or **Pressman '564** or in any combination thereof wherein a reaction mixture is pressurized with carbon monoxide; then heated under pressure of carbon monoxide to a desired ultimate temperature before treatment with carbon monoxide-oxygen mixture as required in steps (iii) and (iv) of claim 34 of the instant application. More particularly, in **Buysch** in Examples where there is a treatment with carbon monoxide before treatment with carbon monoxide-oxygen mixture, the reaction mixture is treated at 60°C with carbon monoxide, then heated to 80-90°C with treatment with carbon monoxide-oxygen mixture (kindly see **Buysch**, Examples 1-4). In Examples in **Mizukami**, the reaction mixture is treated at room temperature with carbon monoxide, then heated to 100°C with treatment with carbon monoxide-oxygen mixture (kindly see **Mizukami**, Examples 1-18). In Examples in **Pressman '272** and **Pressman '564** there is no treatment with carbon monoxide before treatment with carbon monoxide-oxygen mixture.

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In view of these remarks, it is respectfully requested that the rejection of claims 1-56 under 35 USC 103(a) as being unpatentable over **Buysch** and **Mizukami**, or alternatively one of **Pressman '272** and **Pressman '564** be withdrawn.

In view of the foregoing, the Applicants respectfully submit that the application is in condition for allowance. Favorable reconsideration and prompt allowance of the application are respectfully requested.

Should the Examiner believe that anything further is needed to place the application in even better condition for allowance, the Examiner is requested to contact the Applicants' undersigned representative at the telephone number below.

Respectfully submitted,



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